Precipitation Scavenging, Dry Deposition, and Resuspension

Volume 2
Dry Deposition and Resuspension

Proceedings of the Fourth International Conference,
Santa Monica, California, 29 November–3 December, 1982

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DRY DEPOSITION MODEL SENSITIVITY

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INTRODUCTION

Estimates of dry deposition are required to simulate the fate of atmospheric pollutants for a number of important applications. This paper attempts to bound the uncertainty which is likely to exist in current models of plume depletion due to dry deposition. The particular application considered herein is a plume generated by a nuclear power plant accident. We wish to ascertain the bounds on current estimates of deposition velocities for the main effluents of interest. Our analysis will build upon the study by Lewellen and Sheng[1] of the fundamental turbulent interactions within a plant canopy.

We first review our present understanding of the basic dependence of deposition velocity on a number of independent variables. The dependence on atmospheric stability, through its effect on turbulent transport within the surface layer, is the most amenable to direct analysis. The aggregate effect of turbulent transport within individual canopy structures and the viscous sublayer on the basic elements of the surface canopy is considerably less tidy to analyze. The actual surface chemical reactivity can generally only be determined by experiment. If the effluent of interest is in the form of small particles, the viscous sublayer on the canopy surface is likely to be the dominant factor in determining the deposition.

In the final section we attempt to estimate deposition velocity for some of the effluents which may be expected from a nuclear reactor accident. This requires first estimating the uncertainties in the local meteorological variables, the surface canopy variables, and the physical/chemical properties of the main effluents of interest. These uncertainties in the independent variables are translated into uncertainties in the deposition velocity by using the analysis of the previous sections.

REVIEW OF DRY DEPOSITION MODEL

Dry deposition is generally reported as a deposition velocity, V_d, defined as the flux of a gaseous or particulate species divided by the airborne concentration of that species. Physically, it represents the velocity

equivalent to the covariance of the turbulent fluctuations of the vertical velocity and the fluctuations of the species concentration divided by the mean concentration. Since a concentration gradient is generally necessary to deliver a flux to the surface, V_d is necessarily a function of the height at which the normalizing concentration is measured. Since it is a turbulent transport quantity it depends on all of the meteorological parameters governing the wind distribution as well as such diverse phenomena as aerosol dynamics, leaf surface chemistry, plant anatomy, and land use, which determine the surface boundary condition.

Although considerable uncertainty is still involved in several aspects of a general model of dry deposition, we wish to emphasize those aspects which are known and use them to bound the uncertainty which may be expected in dry deposition estimates.

Individual resistances which determine V_d

Deposition of various species in the planetary boundary layer is influenced by the resistances of the various layers through which the species must pass to reach the ground. For ease of depositional analysis, it is convenient to break up the boundary layer into three different regions. The total resistance to deposition, defined as the inverse of the deposition velocity, is then the sum of resistances presented by each region.

$$V_d^{-1} = R_{aerodynamic} + R_{canopy\ film} + R_{surface} \tag{1}$$

$R_{aerodynamic}$ represents the sum of the resistances to turbulent transport in the atmosphere. The second resistance accounts for that resistance due to the thin, viscous, relatively laminar sublayer next to the leaf surface. The third resistance is the surface chemical or biological resistance the species encounters after it reaches the solid surface.

Atmospheric surface layer resistance

For simplicity, we will only consider the constant flux region in estimating the aerodynamic resistance. This should be adequate for the aerodynamic resistance above the canopy, as long as the reference height for defining the deposition velocity does not exceed approximately 100 m.

The similarity solution which exists for the vertical gradients of velocity and species concentration in the constant flux region may be integrated with respect to z to yield expressions for the aerodynamic resistance which are a function of height, z; aerodynamic surface roughness, z_o; stability, L^{-1}; and wind speed. This relationship is plotted in Fig. 1. The resistance is

inversely proportional to the wind speed, so the product of the reference height wind speed and the aerodynamic resistance is presented. At the 1 m reference height, stability has little effect on the resistance, while at 100 m height as much as three orders of magnitude variation is possible. The equivalence between L^{-1} and stability class is indicated for $z_0 = 10$ cm. As given by Golder[2], the stability classes cover a broader (narrower) range of L^{-1} as the surface roughness is increased (decreased).

Canopy film resistance

The canopy introduces source and sink terms into the basic conservation equations for momentum, heat, and species concentration, so the constant flux relationships used to plot Fig. 1 do not hold within the canopy itself. The effective aerodynamic roughness can be used to estimate the atmospheric resistance, but a more detailed analysis of the turbulent interactions within the plant canopy is required to estimate the additional resistance imposed by the viscous air film next to the leaf surface. Lewellen and Sheng[1] developed a model of turbulent flow within a canopy using a second-order closure model of turbulent transport. A key feature of the model is that it distinguishes between the aerodynamic drag imposed by the pressure difference between the upwind and downwind surfaces of a leaf or other object in the canopy, and the skin friction drag associated with the wetted area within the canopy. This distinction is important because the species transfer to the surface is more analogous to the skin friction portion of the drag than it is to the pressure drag. Reference 1 presents some computed results of the sensitivity of the deposition velocity to such variables as the leaf area index, LAI; the ratio of the total wetted area to the projected frontal area, A_w/A_f; the Schmidt number of the species, ν/D; the leaf surface resistance; the wind speed above the canopy; and the atmospheric stability above the canopy.

A major difficulty of such a detailed model is that insufficient data is available to either provide the detailed plant structure functions required for such an approach or to provide the desired model validation. However, the results do suggest a separate factoring of the influences of meteorology, canopy structure, and the physical structure of the species by taking the canopy resistance to be parameterized as

$$R_c U_\ell = R_a (U_h/U_\ell) \, C_s \, P_s \tag{2}$$

where U_h/U_ℓ is the ratio of the velocity at the top of the canopy to that at the reference height, C_s is a canopy structure parameter, and P_s is a parameter depending on the physical structure of the species of interest.

Table 1 provides our guess of C_S for a number of different canopy types. It also includes an estimate of z_o which is needed to determine R_a through Fig. 1, and an estimate of the leaf wetted area per unit horizontal area, LA_wI which will be useful in estimating surface chemical resistance.

TABLE 1: Typical values of characteristic canopy parameters

	LA_wI	C_s	z_o (m)
Grassland	4.	0.5	0.04
Agricultural crop (corn)	20.	0.4	0.2
Suburb	10.	1.0	0.8
Summer deciduous forest	25.	0.2	0.6
Winter deciduous forest	2.	3.0	1.0

A significant deficiency of the analysis of Ref. 1 is that it considered the turbulent production due to drag and buoyancy within the canopy as the sole sources of turbulence. It is known that, under unstable conditions, buoyancy throughout the boundary layer can contribute to the turbulence level at the surface. Thus whenever the characteristic convective turbulent velocity $w_* = [g \, \overline{w'\theta'_o} \, Z_i/T_o]^{1/3}$ exceeds the turbulent shear stress velocity u_* by more than a factor of approximately 2.5, Eq. (2) should be modified to account for this additional source of turbulent transport of species within the canopy. The resultant increase in horizontal wind fluctuations will be essentially passive as far as the vertical transport of momentum or species above the canopy is concerned, but can be quite effective in stirring the air within the canopy to decrease R_c. Without attempting to modify the full model of Ref. 1 to account for this effect, we believe it can be approximately incorporated in Eq. (2) by multiplying it by $[(w_*/2.5u_*)^2+1]^{-1} \simeq [(-Z_i/L)^{2/3}+1]^{-1}$ for unstable conditions.

Effects of physical properties of pollutant species

For the dry deposition of gases in the atmosphere, the physical property of importance that affects dry deposition is the Schmidt number of the gas, $Sc = \nu/D$. The Sc for gases depends principally on the ratio of the molecular weight of the diffusing gas and to that of the medium through which it is diffusing. For diffusion in air, Sc ranges from 0.25 for Hydrogen to between 2.0 and 2.5 for heavier gases like UF6 and Iodine.

The dry deposition of particles is principally dependent on the particle diameter and particle density. The effects of particle density can be roughly incorporated into the particle size by increasing the equivalent particle size by a factor of $\rho^{1/2}$. Particles $\leq$ 10μm in diameter tend to follow all of the turbulent eddies and be diffused in the same manner as a gaseous species, except

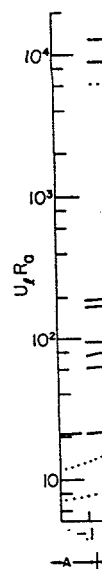opy types. It
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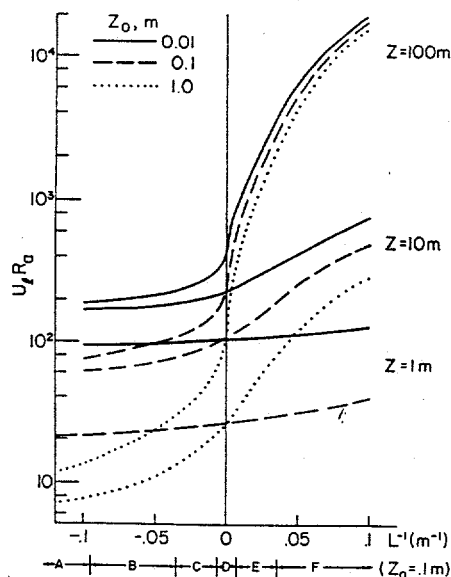can be roughly
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Figure 1. Aerodynamic resistance as a function of stability for three values of aerodynamic roughness and reference height. Stability is measured in terms of the Monin-Obukhov length. The equivalent stability class is indicated for $z_o = 0.1$ m.

Figure 2. The species physical characteristic parameter, P_s, as a function of particle diameter for different values of r.m.s. turbulent velocity q.

in the viscous sublayer next to a surface. Transport in the sublayer is determined by three main contributions – Brownian motion, gravitational settling and inertial impaction. Other possible mechanisms such as thermophoresis, diffusiophoretic forces, electrical migration, etc. may be important under certain conditions but are not included in the current formulation.

The parameter which accounts for the physical structure of the species of interest is modeled here as

$$P_s = \left(\frac{\nu}{D}\right)^{0.7} \left[1 + R_{diff}/R_{imp}\right]^{-1} \tag{3}$$

where, for particles, D is the Brownian diffusion coefficient and R_{diff}/R_{imp} is the ratio of the resistance to Brownian diffusion to that due to inertial impaction. For gases $R_{diff}/R_{imp}=0$. The effect of particle size and turbulence level on P_s is plotted in Fig. 2, as given by the model presented in reference
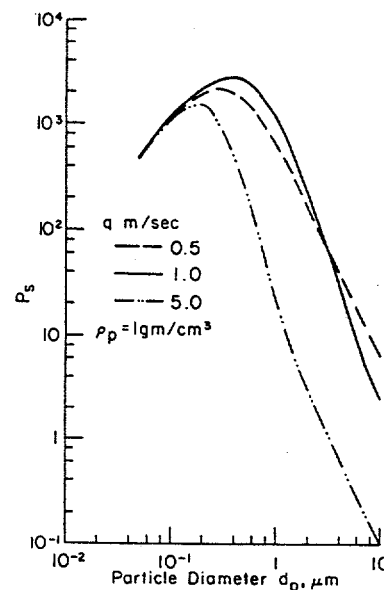
1. This model should be appropriate as long as the characteristic radius of the canopy elements exceeds ~ 0.1 cm, so that the particles are transported to the canopy by the microscale turbulent eddies rather than by direct filtering of the particles not following the larger scale flow around the canopy elements.

Surface chemical resistance

For gaseous pollutants, the surface resistance depends on the chemical reactivity of the gas with the surface material. For plant canopies, the surface resistance also has a biological component, that is, whether the plant stomata are open or closed. With open stomata, the plant "inhales" the pollutant gas and the surface resistance is significantly lower than if the stomata are closed.

The surface resistance needs to be determined experimentally for particular combinations of pollutants and surfaces of interest. Once a data base for R_s has been established, it is possible to estimate the surface resistance for other similar chemical species by extrapolation of the measurements. Garland[3] has reported values of R_s for SO_2 interacting with a number of different surfaces. In general, for soluble reactive gases the R_s will be quite low for wet surfaces and increase by some two orders of magnitude for dry surfaces. The surface resistance for nonreactive gases will be high in general, and not vary much for different surfaces. The surface resistance for the deposition of particles has been considered to be zero in most previous studies, and this assumption has also been used in our current model. In our accounting system any reentrainment processes must be counted as a source term rather than being subtracted f om the deposition.

Within a canopy the R_s which appears in (1) is effectively reduced from the elemental value by the increase in wetted area available for absorption. This may be accomplished by dividing the elemental surface resistance by the wetted area leaf index.

ESTIMATES OF DEPOSITION VELOCITY APPROPRIATE FOR EFFLUENTS FROM A NUCLEAR REACTOR ACCIDENT

The Nuclear Regulatory Commission has established the requirement to consider the deposition of radioactivity up to 50 miles from the power plant site for purposes of evaluating possible ingestion pathways[4]. In the case of an accident, monitoring teams would be sent out to measure actual values of accumulated radioactivity. Accurate models would be an important aid in scoping the possible impact of any specific accident and in providing guidance to a monitoring team.

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Our approach to estimating the appropriate deposition velocity will be to estimate first the physical/chemical properties of the main effluents, which then may be used to estimate the deposition velocity via the model discussed in the previous sections.

Uncertainties in the physical and chemical properties of the main effluents of interest

Previous studies indicate that the principal groups of radioactive effluents expected to be released are: 1) Noble gases - mostly krypton and xenon - that are basically nonreactive; 2) Iodine gas; and 3) Particulate material composed of various radionuclides.

Although the noble gases are nonreactive, and we expect their deposition velocities to be zero because of the essentially infinite surface resistance, it is possible for a noble gas to dissolve in water and thus a wet surface could act as a temporary storage for a noble gas. Since this gas will be released after the plume is past, we believe it is reasonable to consider this more as an added plume dispersal mechanism than it is a true deposition.

The high reactivity of the Iodine gas should act to make its surface resistance quite low. For a wet surface, this surface resistance should be essentially zero, while that for a dry plant leaf is estimated to vary from 0.5 to 5.0 sec/cm, depending upon whether the stomata are open or closed.

The particulate material within the containment vessel will include radionuclides attached to atmospheric aerosols (submicron to a few microns size), particles formed by homogeneous condensation of volatile species (submicron size), larger particles formed by agglomeration of aerosols in regions of high concentration (micron size), water droplets formed in a steam environment using the smaller particles as condensation nuclei (10-20 μm), and some large particles produced due to possible fuel rod rupture (10-100 μm). A core melt sequence is expected to lead to the formation of large quantities of aerosols due to concrete decomposition composed of silicon and calcium with attached radionuclides. According to reference 5, the particle size is expected to be 2 μm aerodynamic mass median diameter with a geometric standard deviation of 2 for a log normal distribution. This corresponds to most of the particles ranging from 0.5 μm diameter to 10 μm diameter. The density of these particles will range from atmospheric aerosol densities to heavy element particles with densities of as much as 20 gm/cm^3. The particle size distribution out in the atmosphere from failed containments will be at least as broad as that within the containment vessel. Thus, from Fig. 2, P_s can range from 1 to 10^3.

Uncertainties in the meteorological and surface variables

In developing an emergency response model, it is not possible to know in advance what the meteorological conditions will be at the time of any accident. In assessing possible impact it is therefore prudent to assume "worst-case meteorology". However, it is not clear what conditions would constitute the worst case since conditions which reduce the deposition relatively close to the release point lead to increased deposition at a distance. Which is worse depends on the land-use pattern around the specific plant. It follows that a wide range of meteorological conditions should be considered to permit the model to estimate the worst case.

One situation which is likely to lead to large deposition relatively close to the source is the early part of the morning transition from stable to unstable atmospheric conditions after a night with heavy dew. The remnant dew will serve to effectively eliminate any surface resistance for the iodine gas while the decreasing atmospheric stability acts to reduce the aerodynamic resistance. Also the relatively low mixed layer height at this time will keep a low level release relatively close to the surface. The actual wind speed does not appear to be very important in determining the total deposition. Both the deposition velocity and the plume transport velocity are roughly proportional to the wind speed. Thus the fraction of a plume deposited within a given distance of the source should be relatively independent of wind speed.

The worst case for deposition at an appreciable distance from the source is likely to be associated with a release which occurs a couple of hours earlier in the morning. In this case, the high stability of the atmosphere can act both to hold down the deposition velocity in close, and permit the plume to travel some distance without wide dispersal. When the morning transition from stable to unstable occurs it then permits relatively heavy deposition of the plume at an appreciable distance from the release point.

The full range of stability conditions need to be considered when assessing the impact of future accidents, but the stability input to the deposition velocity estimate should be compatible with the stability choices used in the dispersion calculation.

Resultant uncertainty in deposition velocity

Fig. 3 shows the result of applying the ranges discussed in the last two sections to the analysis represented by Figs. 1 and 2. The combined result represents approximately 5 orders of magnitude spread in the ratio of the deposition velocity to the wind velocity. If allowance is made for one order of magnitude variation in the wind speed, this yields a probable range of 6 orders
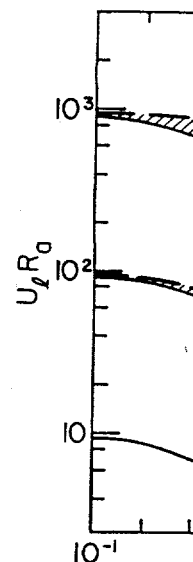

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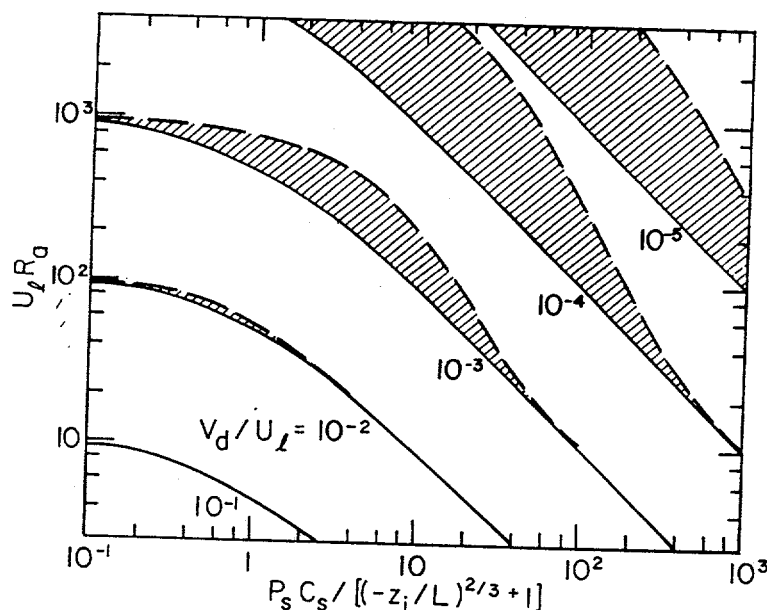most amenable to an...
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Figure 3. Contours of constant deposition velocity as a function of aerodynamic resistance, R_a; reference layer wind speed, U_ℓ; the species physical characteristic parameter, P_s; and the canopy structure parameter, C_s. Shaded area represents variation in U_h/U_ℓ. The parameter involving the mixed layer height, Z_i and the Monin-Obukhov length, L is to be used under unstable conditions only. The surface resistance is zero for this plot.

of magnitude between the deposition velocity associated with an Iodine plume for an unstable atmosphere in a wet forest canopy, and that associated with a plume of particles of 0.2 μm diameter size in a stable light wind above smooth grassland. Certainly, using a simple "representative value" of 1 cm/sec as the deposition velocity in an analysis to assess the possible impact of a plume can lead to erroneous conclusions. For engineering purposes a value of $V_d/U_\ell \leq 10^{-4}$ may be taken as zero when downwind distances of 100 km or less are considered. The range of uncertainty of interest for the present problem then is 3 orders of magnitude.

A substantial part of the resultant uncertainty is associated with the various possible atmospheric stability conditions, particularly when the deposition velocity is referenced to 100 m. However, this sensitivity is the most amenable to analysis as illustrated in Fig. 1. When the deposition velocity is to be used in conjunction with a dispersion model calculation then, at least, the rough stability class will be specified for use with that

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dispersion model. This allows the choice of R_a to be correspondingly narrowed. The thickness of the model layer adjacent to the ground dictates the z height which should be used in Fig. 1. If a simple Gaussian plume model is to be used, then a height characteristic of the plume is appropriate. Thus, stability will introduce much more uncertainty into estimates of V_d for use with single layer plume models than in detailed numerical models which carry a relatively large number of vertical layers with a thin surface layer.

When the meteorology of a particular time is considered, then the largest uncertainty is likely to be associated with the uncertainty involved in the distribution of particle sizes released. As may be seen from Fig. 2, narrowing the range of particle sizes rapidly decreases the uncertainty in P_s. On the other hand, when the canopy is lush so that $C_s \lesssim 0.2$ and typical afternoon conditions are considered, so that $L/Z_i \approx 10^{-2}$, then Fig. 3 will yield a value of V_d for submicron particles with $P_s = 10^3$ which is within one order of magnitude of that obtained for larger particles with $P_s = 1$.

CONCLUDING REMARKS

Our analysis provides an estimate of the uncertainty existing in current estimates of the deposition velocity as a result of uncertainties in certain independent variables such as canopy structure, surface-layer stability, particle size, and surface chemical reactivity. It indicates that all of these variables need to be determined if a relatively accurate estimate of V_d is desired for a particular time and place. It also suggests that it is possible to ascribe much of the data scatter reported in reviews such as Sehmel's[6] to uncertainty in some of these key experimental variables. Although some of the uncertainty in deposition velocity estimates may still be attributed to modeling deficiencies, particularly as related to mechanisms other than turbulent inertia or Brownian motion for transporting micron size particles across the viscous sublayer, much of it can be directly attributed to uncertainty in key independent variables.

ACKNOWLEDGMENTS

We appreciate the support of this work by the Nuclear Regulatory Commission with R.F. Abbey Technical Monitor. We also acknowledge useful comments by W.G.N. Slinn and K.T. Paw U. in their review of an earlier version of this paper.

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DISCUSSION

W.G.N. SLINN: 1. For particles whose gravitational settling is important, the resistance analogy used by the authors is invalid.

2. When particle inertia is significant (and possibly also for interception), the authors' use of Reynolds' analogy (ignoring form drag) is invalid.

3. In Eq. (3), interception has been ignored; yet Chamberlain's data show interception's significance.

4. Fig. 2 does not show the "inertial hump" at the larger wind speed, and apparently does not contain interception; it is therefore quite inadequate.

5. For iodine released from nuclear facilities, the authors should have commented on iodine's chemical form, the photolysis of CH_3I (and the possible photolysis of I_2), and attachment of the gases and ions to released and ambient aerosol particles.

6. In contrast to the authors' statement, it is common that if the particle size distribution is narrowed, then the uncertainty in V_d increases (because errors are no longer averaged).

7. If interception and the polydispersity of aerosols is included, then the uncertainty range is reduced from that described by the authors.

8. The reader could have been helped if the references had given a better reflection of current knowledge about dry deposition.

W.S. LEWELLEN, A.K. VARMA and Y.P. SHENG: 1. Gravitational settling imposes a lower bound on V_d. Above this lower bound the division into separate resistances is still very useful since particles of 10 μm or less will generally follow the turbulent eddies except in the thin viscous layer adjacent to the canopy elements.

2. This is the reason for including the parameter R_{diff}/R_{imp} in Eq. (3), and for distinguishing between form drag and friction drag in the basic model of the canopy.

3. What you refer to as interception must be included in the R_{diff}/R_{imp}. It is true that R_{diff}/R_{imp} should depend on the characteristic radius of canopy elements when curvature of the flow around canopy elements exceeds the curvature imposed by the microscale turbulent eddies. This will occur when the characteristic radius is less than approximately 0.1 cm.

4. Disagree. Figure 2 does include the inertial hump. It was not evident in the figure provided with the review copy but is quite distinct for the largest value of q in the present figure.

5. We agree that plume chemistry can greatly compound the problem, but is unlikely to reduce the range of uncertainty in V_d.

6. Disagree. Broad particle size distribution means size distribution may change as a function of downwind distance which is unlikely to reduce uncertainty. It is true that a broader size distribution will under a number of conditions yield a larger, easier to measure value of V_d, but this should not be equated with saying the broader distribution provides a more certain estimate of V_d.

7. As noted earlier, we have attempted to include interception in our analysis and as noted in the last comment, only partially accept that the polydispersity of aerosols reduces the uncertainty. Thus we believe, as stated in the paper, that a range of 3 orders of magnitude variation in deposition is appropriate when assessing the impact of a nuclear reactor accident.

8. We agree, but in order to have included survey material within our allotted space, we would have had to streamline our basic argument much more. Our Ref. 1 includes a reference list of 46 publications for those who wish to pursue the details and foundation of our analysis. Since this paper is intended to appear in a book accompanied by a large number of other dry deposition papers, we felt confident that readers would receive a good reflection of current knowledge about dry deposition even if we short-changed them a bit in our particular article.

M.L. WESELY: The variability of wind speeds, especially the peak gusts, inside canopies might not always "scale" well with u_* or $u_\ell C_D^{1/2}$ alone. Some dependence on atmospheric stability or depth of mixed layer (in unstable conditions) seems possible. Might diffusion, impaction, and interception of particles to individual leaves be considerably enhanced in such conditions?

W.S. LEWELLEN, A.K. VARMA and Y.P. SHENG: True, the turbulence level within the canopy should not be expected to scale with u_* whenever u_* is appreciably less than the characteristic convective velocity w_*, just as the horizontal velocity fluctuations in the constant flux layer above the canopy do not scale with u_* under the same conditions. As a result of my discussion with you at the conference we have attempted to include this effect on page 4 in the present version of the paper. This significantly increases the model's expected value of deposition for 0.5 μm size particles under afternoon conditions.